CHAPTER 7

Conclusions & Future Work
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7.1 Conclusions

The important conclusions of this PhD research are summarized in the sections below as:

7.1.1 Conclusions for Experimental Results of EFC Process

The role of molecular architecture of entangled flexible polymers on their necking behavior during steady state extrusion film casting was interrogated. Specifically, the effects of long chain branching (LCB) and polydispersity on necking was interrogated. Experimental studies were performed on four commercial polyethylene samples: a linear PE of low polydispersity (LLDPE 2045G), a linear PE of high polydispersity (HDPE DMDH6400), a sparsely long chain branched PE (m-LLDPE PL1840G) and a PE containing significant amounts of long chain branching (LDPE 170A). All polymers showed increasing necking with increase in DR and TUL. For a given draw ratio and take up length the extent of necking decreased in the order 2045G > PL1840G > DMDH6400 > 170A. These observations are in agreement with those reported in recent literature.

Polydispersity is an important molecular attribute in determining the extent of necking for linear polymers. We observed that necking was substantially reduced (for fixed DR and TUL) for the broad MWD containing HDPE DMDH6400 vis-à-vis LLDPE 2045G that had narrow MWD. The broad MWD combined with high \( M_z \) fraction for the HDPE resin was responsible for reduced necking since the presence of these macromolecular attributes gave rise to extensional strain hardening that resisted necking.

Experimentally, several differences were observed between the linear LLDPE 2045G and the branched LDPE 170A. For a given TUL, both LDPE and LLDPE undergo necking and the extent of necking increases with DR. The
branched LDPE necks to a lesser extent than the linear LLDPE at a given DR and TUL. Consequently, the LDPE film thins down to a greater extent as compared to the LLDPE film at equivalent DR and TUL. Additionally, for both resins, the extent of necking increases with increase in TUL at a fixed DR with the linear LLDPE displaying greater necking than the branched LDPE. Thus, the presence of long chain branching (LCB) (as present in LDPE) reduces necking and enhances the drawdown of the extruded cast films when compared with the linear resins. These observations are independent of the way in which the draw ratio is set.

For similar DRs, the velocity of the LLDPE film close to the die exit is lower than that for the LDPE and remains lower throughout the take-up length. Both polymers showed a small region of extrudate swell near the die exit. This shows up as a small decrease in the measured velocity. Beyond this region, the lower velocity of LLDPE near the die exit is because of the larger width of the film in this region. At higher DR, the centerline velocity for LLDPE showed a somewhat exponentially increasing profile while the LDPE showed a linearly increasing profile. This trend was qualitatively similar to that observed by previous researchers who attributed this behavior to increasing viscoelasticity of the melt.

In general, it was observed that the transverse velocity profiles for both LDPE 170A and LLDPE 2045G was reasonably close to being linear, which was one of the principal flow kinematics assumptions while modeling the EFC process. From the experimentally determined transverse velocity profiles for LLDPE 2045G and LDPE 170A, it was observed that at low axial positions (near the die exit), at equivalent DR's, the branched LDPE always displayed higher $V_y$ velocities at the outermost edge (labeled lower edge) as compared to linear LLDPE. This higher $V_y$ was responsible for enhanced necking of LDPE just near the die exit vis-à-vis the linear LLDPE. Near to the chill-rolls, the $V_y$ values for LLDPE were consistently higher than those for the LDPE indicative of higher necking for LLDPE at the chill-rolls vis-à-vis LDPE.
For both LDPE 170A and LLDPE 2045G polymers the temperature drops from about 190°C (463 K) at the die exit to about 100°C (373 K) at the chill-rolls with a higher temperature gradient near the die exit. For both films the temperature drops to larger extent for higher draw ratios. The greater extent of cooling for higher draw ratio observed experimentally clearly suggests the dominance of convective heat transfer over other mechanisms of cooling under the experimental conditions prevalent in this work. It was observed for the higher DR that the LDPE film cools to a greater extent than the LLDPE film, especially in the region near the die exit. The lower temperature of the LDPE film near the die exit can be attributed to its higher surface velocity compared to the LLDPE film. This causes greater cooling of the LDPE film under the convection-dominated heat transfer regime of our experiments.

Finally, prominent edge-beading was observed for all PE films made by the EFC process at all DR’s and TUL’s. While it was observed that the branched LDPE 170A followed Dobroth-Erwin scalings for almost all DR’s and TUL’s, the linear LLDPE 2045G followed the scalings only at the lowest TUL of 10 mm.

### 7.1.2 Conclusions for Isothermal CFD Simulations of EFC Process

The effect of molecular architecture on necking was further studied by modeling the film casting process using a 1-D isothermal flow model, and incorporating into it two multi-mode molecular constitutive equations: the Rolie-Poly-Stretch (RP-S) equation for linear polymers and the eXtended Pom-Pom (XPP) equation for branched polymers. The model predictions of the effect of DR and TUL on necking were found to be in qualitative agreement with our experimental data. The model also relates the observed differences in film widths of linear and branched polymers to chain stretching, thereby providing molecular basis to the macroscopic necking phenomenon. Additionally, the model predicts the following four key results all of which are in agreement with recent experimental reports: (1) The
presence of sparse long chain branching reduces necking at smaller draw ratio, but does not affect the necking at higher draw ratio, (2) The presence of large number of long chain branching substantially decreases necking at all draw ratios, (3) The presence of long chain branching on faster modes of branched polymer causes stronger necking closer to the die but reduced necking downstream towards the chill roll, and (4) the increase in polydispersity, and specifically the presence of very long chains in the molecular weight distribution of linear polymers, results in reduced necking.

The model fails to provide quantitative comparisons with experiments because of the assumptions made in this minimalist approach, such as the 1D flow kinematics and isothermal conditions. Indeed, temperature measurements made in our experiments have shown significant cooling of the film for larger take up lengths of 90 and 230 mm. Similarly, the experimentally observed necking profile of LLDPE, which shows a region of little necking close to the die, is not predicted by the 1-D flow model. Finally, the 1-D flow model cannot predict edge-beading profiles because of its intrinsic assumption of constant film thickness for fixed axial position.

Nevertheless the use of molecular models such as the RP-S and the XPP in a 1D flow model enables the understanding of the role of relaxation processes on the necking phenomenon, and provide useful guidelines to design polymer grades to meet demands such as reduced necking in the film casting process.

### 7.1.3 Conclusions for Non-isothermal CFD Simulations of EFC Process

The second-generation 1-D flow model, which includes molecular constitutive equations, energy equation and a temperature dependent viscosity and relaxation spectrum of Arrhenius form, predicts the film dimensions better than the first-generation isothermal model. The model also provides better predictions of centerline velocity and temperature profiles. The model relates the shape of the predicted necking profile to the molecular stretch. The key difference between the non-isothermal and
isothermal predictions is that the film necks in and draws down closer to the die; both the width and thickness tend to saturate near the chill roll. This is because the film resists deformation as it cools down. The presence of long chain branching, leading to strain hardening under extensional flow causes further increase in resistance to deformation. Thus an increase in branching causes the film to further neck-in and draw down earlier near the die-exit.

The non-isothermal predictions showed favorable comparison with experimental data vis-à-vis isothermal predictions. There was a clear difference in the film velocity in axial direction \( V_x \) between isothermal and non-isothermal model predictions. Under non-isothermal conditions, the ability of the film to resist deformation increases as it cools down during its travel to the chill roll. Since the axial velocity has to reach the set value at the chill roll, the film prefers to accelerate earlier in the region where it is relatively hotter, i.e. near the die exit. On the other hand, under isothermal conditions, the film can afford to deform later, i.e. near the chill roll. Therefore, the axial velocities predicted by the non-isothermal model were higher than those predicted by the isothermal simulation for the same DR. For the transverse velocity \( V_x \) profiles for the linear LLDPE and branched LDPE resins, the numerical predictions show qualitative agreement with experimental data at low DR (of 4) but not at high DR (of 17).

There are several limitations associated with the 1-D flow kinematics that do not allow for a full quantitative comparison with experimental data. Additionally, extrudate swelling is unaccounted for in this analysis which can play an important role in computing the final shape of the necking profile. However, the non-isothermal 1-D model with molecular constitutive equations provides useful insights into the various effects of molecular structure on film necking.
7.2 Recommendations for Future Work

The following are recommendations for future work in this area:

1. Undertake 2-D (or quasi 3-D) finite element (FEM) simulations to quantitatively predict both necking and edge-beading under both isothermal and non-isothermal conditions.

2. Undertake CFD simulations of flow inside the cast film die so as to obtain better predictions of die exit conditions such as stresses and extrudate swell.

3. Utilize a rheo-optical flow birefringence set-up built around an EFC facility to measure the stresses in the molten polymer film that are responsible for necking and edge-beading phenomena.

4. Build upon the understanding developed in this research to investigate EFC for biodegradable polymers such as PLA and other polymers such as LCB containing PP.